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# Charge Distribution of $^{16}O$ Ions Produced from Tandem Accelerator*

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In an effort to obtain information about the acceleration of heavy ions in tandem accelerators, the charge state distribution of oxygen ions accelerated in the University of Washington FN tandem accelerator was measured at three terminal voltages and over a range of stripper pressures. It is found that the stripper pressure normally used is below the value required for equilibrium in the charge exchange process, but that at higher terminal voltages the average charge state of the ions can rise above the equilibrium value due to focusing effects in the accelerating tubes. This phenomenon has implications in the use of foil strippers with heavy ion beams. A useful empirical relation for the optimum stripper pressure for a given charge state is obtained.

HEAVY ions have been accelerated in tandem Van de Graaff accelerators for some time now and have proved extremely valuable as nuclear probes.[1] Although some information is available on the ionic charge exchange and stripping of such ions at tandem energies in circumstances of good geometry,[2] there seems to be very little available data on the more practical matter of their behavior inside a tandem accelerator. Information of this kind is of value because it can lead to a better understanding of the processes affecting the production of heavy ion beams and suggest directions for development and improvement. In an effort to obtain such information, a study of the charge state distributions of $^{16}O$ ions accelerated in the University of Washington FN tandem Van de Graaff accelerator[3] was undertaken. Charge distributions were studied as functions of terminal potential and stripper gas pressure. Oxygen ions were selected for this study because they are relatively easy to produce and can form enough different charge states to show clearly the shape of the charge-state distribution.

A beam of approximately 2 $\mu$A of negative oxygen ions was obtained from the standard negative ion source, using the Rutgers technique[4]: The duoplasmatron, in which had been installed a fresh barium oxide coated, platinum mesh filament, was operated on hydrogen gas. The filament coating introduced enough oxygen into the plasma discharge to provide an output of oxygen ions, and this output was maximized by adjustment of the source parameters. The negative oxygen ion beam was introduced into the accelerator, an FN tandem equipped with inclined field accelerating tubes of the Mark I design[3] at the low energy end of the machine and Mark II tubes[3] at the high energy end. Oxygen was used as the stripper gas. The stripper tube was 83.8 cm long and 6.8 mm in diameter.

Two methods of studying the ionic charge distribution were employed. The first method involved separate magnetic analysis of each charge state present at a given terminal and stripper pressure. The analyzing magnet was set for a field which would bend the charge state under study through 90°, so that the corona regulation system would control on this beam. The analyzed beam current, as read in a deep Faraday cup, was then maximized by adjusting the focusing and steering at the output of the machine. In this way the absolute yield of each charge state capable of controlling the machine was determined.

This method, while permitting accurate measurement of the oxygen charge state distribution, proved rather time consuming. In addition, it suffered from other difficulties. The major of these was the inability to observe charge states too weak to operate the corona stabilization system. This problem was particularly severe with charge states 1 and 2, which are usually of low intensity and carry less charge (and therefore current) per particle to the control slits. Further, the lower charge states, particularly at high energies, required magnetic fields outside the range of our NMR field measuring instrument, thus making the location of the lower charge states difficult and somewhat uncertain. Finally, since the charge-state distribution measurements were made point-by-point, they were vulnerable while the measurements were in progress to slight changes in tube pressure, ion source output, or other machine parameters which might alter the charge distribution or over-all intensity.

For these reasons, a second method of determining the ionic charge distribution was also used. The focusing and analyzing magnets and the steering electrodes were turned off, and the beam emerging from the tandem accelerator was defined by 5.08 mm vertical and horizontal object slits. The compensating coils of the analyzing magnet were employed to balance out any residual field left in the iron, so that the beam could pass straight through the magnet without being deflected. Visual inspection of the beam on a quartz plate about 12 m from the accelerator showed no

* Work supported in part by the U. S. Atomic Energy Commission.
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[1] H. E. Gove, Nuclear Instr. Meth. **11**, 63 (1961); P. Rose, K. H. Purser, and A. B. Wittkower, IEEE Trans. Nucl. Sci. **NS-12**, No. 3, 251 (1965).
[2] E. Almqvist, C. Broude, M. A. Clark, J. A. Kuehner, and A. E. Litherland, Can. J. Phys. **40**, 954 (1962).
[3] Manufactured by High Voltage Engineering Co., Burlington, Massachusetts.
[4] J. DeBoer, Rutgers University (private communication).

visible dispersion of the ion beam, although considerable dispersion could be produced and several charge states observed by introducing a small magnetic field in the analyzing magnet.

A small scattering chamber was mounted at the 0° (undeflected) port of the analyzing magnet and the $^{16}O$ beam was passed through a defining aperture 6.35 mm in diameter before striking a 150 $\mu g/cm^2$ gold target 15 cm behind the aperture. Oxygen ions Rutherford scattered through an angle of 90° in transmission by the gold target were passed through a pair of defining slits 1.57×4.75 mm separated by 5.08 cm and detected by a diffused junction silicon, charged particle detector. Figure 1 shows the experimental geometry used in this experiment.

Since the various charge states of oxygen were accelerated to different discrete energies by the tandem, the particle detector produced a pulse-height distribution showing prominent groups corresponding to the different charge state components of the incident beam. This technique permits observation of the relative intensities of all charge states at once, and avoids ambiguities arising from uncertainties in the magnetic analysis system and from small fluctuations in beam energy.

Since the ions were scattered from gold before being detected, their energy distribution was modulated by the energy dependence of Rutherford scattering, which goes as the inverse square of the energy. This modulation has the effect of enhancing the lower charge states in the observed energy distribution while reducing the contribution of the higher charge states. This provides built-in compensation for the low charge states, which are of rather low intensity, but at the same time makes it difficult to observe the very highest charge states, which are of great practical interest since they have the highest energies.

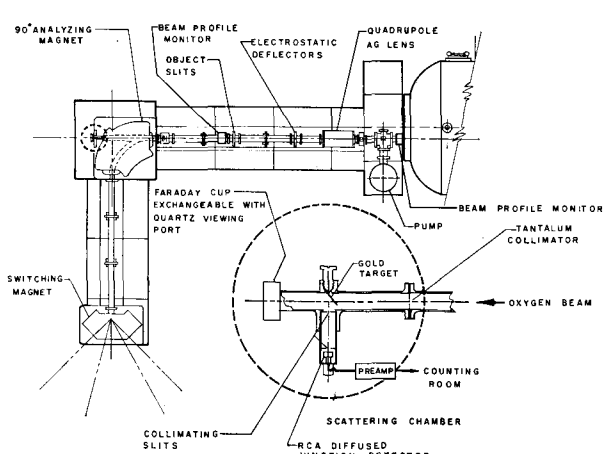


FIG. 1. External beam system of University of Washington FN tandem Van de Graaff accelerator showing position of scattering chamber.

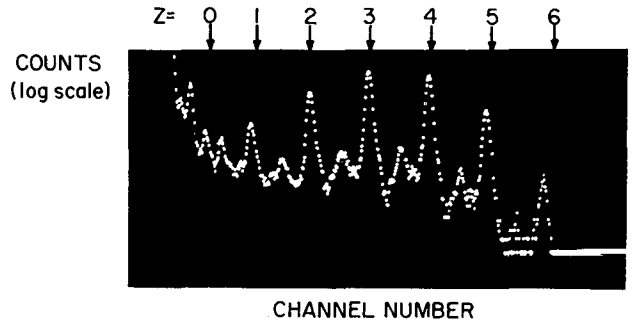


FIG. 2. Pulse height spectrum observed with 6.5 mV terminal potential. Arrows indicate positions of charge state peaks.

The stripper gas pressure was then varied, and pulse height spectra from the particle detector were recorded with a 512 channel multichannel analyzer at terminal potentials of 2.5, 4.5, and 6.5 MV and over a range of pressures.

Figure 2 shows a typical pulse height spectrum obtained in this way. Peaks corresponding to $^{16}O$ charge states zero (neutral) through six are present in the distribution. Curiously, there are also a smaller set of peaks half way between the main charge state peaks. These "half integral" peaks were at first attributed to a dead layer on the detector, but their regular spacing, uniform resolution, and energy dependence rule out this possibility.

It is now believed that they arise from conversion of $O_2^{--}$ ions from the ion source to $O_2^{-}$ ions through collisions with residual gas molecules in the beam tube between the ion source and the entrance to the accelerator. This suggests that a significant fraction of the negative oxygen ions produced by the ion source are not $O^-$ ions, as is usually assumed, but $O_2^{--}$ ions which are electrically identical to $O^-$.

No means of measuring directly the pressure within the stripper tube was available, and so the reading of the high energy vacuum gauge was taken as an index of stripper pressure. To establish the relation between this gauge reading and the actual areal density of stripper gas which the ion beam encounters, a calibrated leak was employed to determine the response of the gauge to a given gas flow rate. It was found that a leak of 5 atm $cm^3/h$ produces a gauge reading of $1.0\times10^{-5}$ Torr. Using this information, the dimensions of the stripper tube, and molecular flow pumping speed tables for long tubes[5] it was calculated that a H.E. vacuum gauge reading of $10^{-5}$ Torr corresponds to a stripper gas areal density of approximately 0.5 $\mu g/cm^2$ or about $1\times10^{16}$ atoms/$cm^2$. Since there is some uncertainty in the accuracy of these values, the actual vacuum gauge reading was retained as the stripper pressure index in the presentation of the measured charge distributions. These distributions, measured over a range of stripper

[5] Saul Dushman, *Scientific Foundations of Vacuum Techniques* (John Wiley & Sons, Inc., New York, 1962), 2nd ed., Ch. 2.

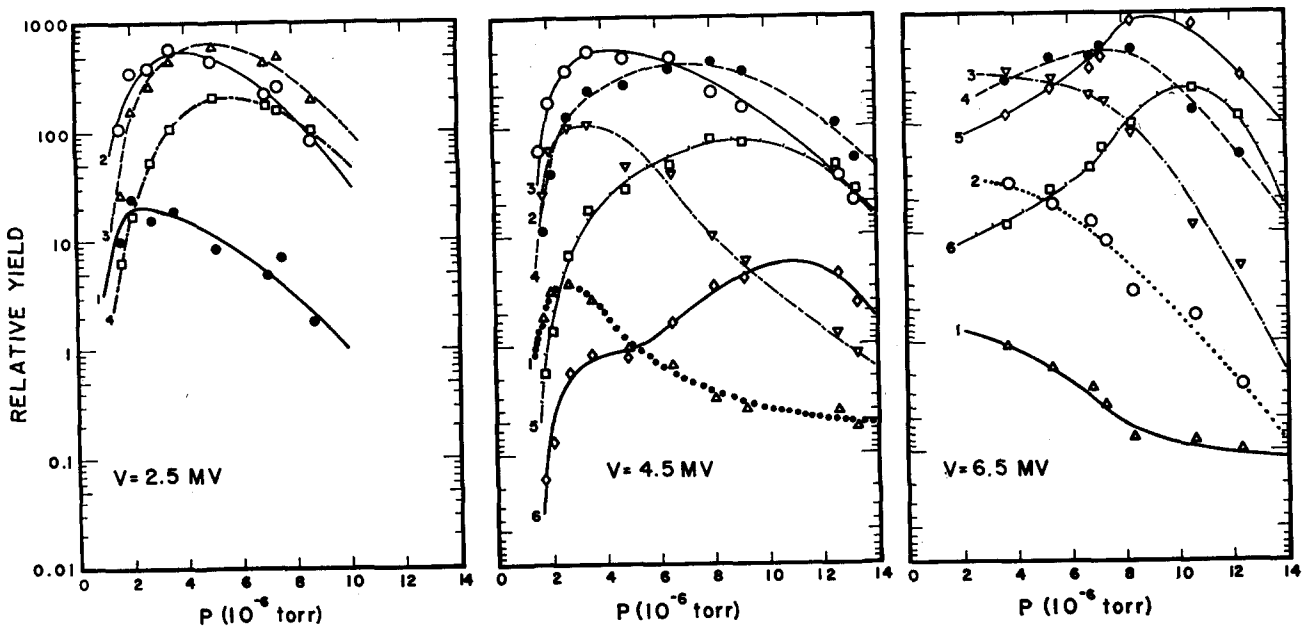


FIG. 3. Observed yield of multiply charged ions as measured with apparatus shown in Fig. 1 over a range of stripper pressures at three terminal potentials. The small numerals at the left end of the curves indicate the charge of the ions.

pressures at terminal voltages of 2.5, 4.5, and 6.5 MV, are shown in Fig. 3.

The relative errors arising from counting statistics and estimation of background in the scattering measurements were 3% or less. Absolute error was a more serious problem and had two primary sources—uncertainties in determining the absolute intensities of charge states with magnetic analysis, and possible dispersion effects during the scattering measurements arising from the characteristics of the accelerating tubes or residual fields in the unused quadrupole lens. These errors are estimated to be no greater than 20%. It should be noted in this context that the agreement between relative intensities obtained from scattering and from magnetic analysis measurements was always within 5%.

In addition to these errors, some uncertainty is associated with the pressure values of the data points. This arises from readoff errors in reading the high energy vacuum gauge, possible calibration errors of this gauge, and from the relatively long time ($\sim$10 min) required for the stripper pressure to reach equilibrium. Errors in abscissa value are estimated to be less than $\pm 10\%$.

To gain some understanding of the results of these measurements it is necessary to consider the four principal effects which combine to produce them:

(a) The average charge of the ions produced by the stripper increases with the stripper gas pressure inside the stripper tube until an equilibrium in the charge exchange process has been established. We note that 8.7 MeV $^{16}O$ ions reach equilibrium in argon gas when the areal density of stripper gas is approximately $3\times10^{16}$ atoms/cm$^2$, and that the charge-exchange cross section for argon is expected to be somewhat larger than that of oxygen.[6,7] This leads us to the conclusion that the oxygen ions have probably not reached the equilibrium charge distribution at the stripper pressures used. This conclusion is in agreement with Fig. 4, which shows the average charge $Z_{av}$ at various pressures as a function of energy, and indicates that in general the average charge falls below the expected equilibrium value.[6]

(b) At a given stripper pressure either at or below equilibrium, the average ionic charge increases with energy. This expectation is also confirmed by Fig. 4.

(c) The ions, in colliding with stripper gas atoms, suffer small angle scatterings. Some of these scatterings are through angles sufficient to cause the ions to collide with the walls of the stripper tube. The fraction of particles lost from the beam is essentially independent of charge state, but is proportional to stripper pressure and inversely proportional to the square of the energy of the ions. Thus, this effect does not alter the charge-state distribution, but may, when the stripper pressure is sufficiently high, result in substantial loss in beam intensity. The angle at which such losses become serious is approximately 0.5° for the present stripper tube. The fact that such small-angle scatterings can be of considerable importance has been shown at this Laboratory in work involving 9 MeV $^{12}C$ ions.[8]

(d) The divergence produced by small-angle scattering in the stripper can also result in loss of beam in the

[6] E. L. Hubbard, Ann. Rev. Nucl. Sci. 11, 419 (1961).
[7] L. C. Northcliffe, Ann. Rev. Nucl. Sci. 13, 67 (1963).
[8] E. Preikschat and R. Vandenbosch (to be published).

accelerating tubes by collisions with the tube electrodes. Since the accelerating tubes tend to refocus this diverging beam, and since the strength of such focusing increases with the charge state of the ion, this effect can result in better transmission for high charge states than for low. This effect is not expected to alter the average ion charge state unless (1) there is substantial loss of beam due to collisions with the tube electrodes and (2) there is a large enough fraction of the higher charge states in the ion beam so that increased transmission for these ions has a significant effect on the average charge state.

With the above considerations in mind, let us examine Fig. 4. As expected, the average charge state increases with increasing stripper gas pressure and increasing terminal potential. The $Z_{av}$ curves at the lower pressures have a smaller slope than the equilibrium-charge curve. This is in agreement with the calculation mentioned earlier which shows that even when $p=10^{-5}$ Torr the stripper gas areal density is only about $10^{16}$ atoms/cm$^2$, well below that required for equilibrium in the charge exchange process. A given stripper pressure more nearly produces an equilibrium charge distribution at low energies than at high. Thus as the energy is increased the average charge, although increasing, can be expected to fall further below the equilibrium value, and the slope should be less than that of the equilibrium charge energy.

The most striking feature of Fig. 4, however, is the behavior of the average charge at higher stripper pressures. Here the average charge rises sharply at 6.5 MV, and even *exceeds* the value of the equilibrium charge at this energy. This is interpreted as a manifestation of effect (d) discussed above. The small-angle scattering, which goes as $E^{-2}$, should be smaller at this energy than at the lower energies studied. Moreover, there is a significant fraction of charge states 5 and 6 at this energy. Thus, as the small angle

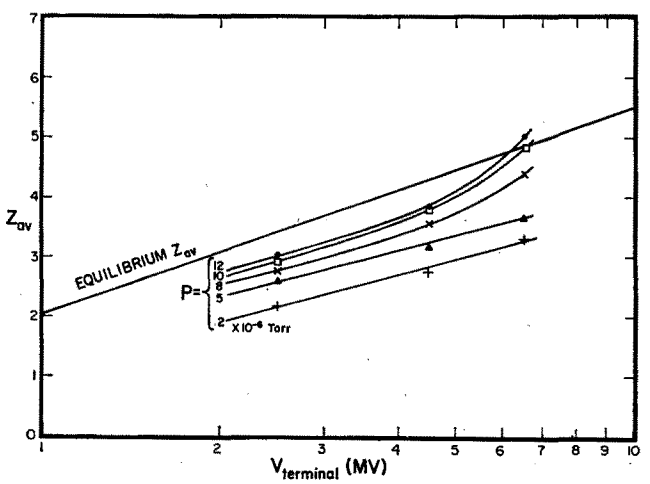


FIG. 4. Average charge computed from measurements shown in Fig. 3 as a function of terminal potential and stripper pressure. Calculated average equilibrium charge is shown for comparison.

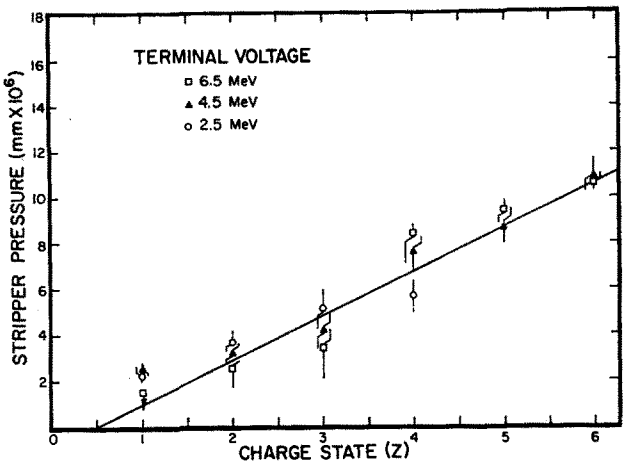


FIG. 5. Optimum stripper pressure to give maximum yield of a particular charge state at three terminal potentials. Flags indicate pressure range over which 90% of maximum yield is maintained.

scattering falls and the higher charge states appear, a condition is reached at which the refocusing of scattered ions of high charge begins to play an important role in the determination of the average charge. This explains the rise of $Z_{av}$ at 6.5 MV and the fact that it exceeds the equilibrium value.

These data were also analyzed in terms of the stripper pressure at which a maximum of a given charge state was obtained. These results are indicated in Fig. 5. The error bars indicate the width of the peak and show the pressures at which the yield falls to 90% of the peak value. We see that the optimum stripper pressures indicated by these data are relatively independent of energy and are fairly well represented by a straight line. This provides a useful rule-of-thumb for setting up the accelerator to produce oxygen in a given charge state, and this type of behavior would be expected in other kinds of ions as well.

We have seen from the above discussion that the maximum transmission of oxygen ions in an FN tandem accelerator takes place at stripper gas pressures below that required for charge exchange equilibrium. Operation at higher pressures is not feasible because of intensity loss due to small-angle scattering, although some of these losses were reduced by refocusing of ions in the higher charge states. The ions which are refocused must have had small enough angular divergence to avoid collisions with the stripper tube walls, yet would have been lost by collisions with tube electrodes if they had not been refocused. This implies that the accelerating tube places a tighter restriction on the angular divergence of the stripped ion beam than does the stripper tube itself. Thus it is doubtful if increasing the aperture of the stripper tube would have a sizable effect on the transmission of the accelerator. On the other hand, providing some strong focusing device at the terminal just after the stripper, perhaps a permanent magnet quadrupole, might provide a substantial increase

in the transmission of oxygen ions and allow the use of higher stripper pressures so that a higher average charge could be achieved. It should also be noted that there has been considerable discussion of the use and relative merits of foil strippers in tandem accelerators. Since almost any reasonable foil is at least 5 $\mu g/cm^2$ thick, about an order of magnitude larger than the estimated gas thicknesses used here, the use of such strippers without some strong focusing at the terminal would be expected to greatly reduce the output of oxygen ions because of scattering losses in the accelerating tube.

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# Experimental Considerations for Polaroid Film X-Ray Photographs

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Polaroid type 57 sheet film (ASA 3000) has been evaluated as a means for recording x-ray Laue and pinhole diffraction photographs; both transmission and back reflection geometries have been employed. Consideration has been given to the effect of x-ray tube target material, tube voltage and current, exposure and processing times, collimator size, and specimen-to-film distance on resultant pattern quality. Examples are given which illustrate the the ability to obtain high quality Polaroid film patterns in considerably less (5 to 20%) time than is required for comparable wet process films.



## INTRODUCTION

THE application of Polaroid Land film to x-ray diffraction studies has been shown to result in a considerable reduction of exposure and processing times[1–4] and must therefore be considered as a practical and economical method for routine laboratory and production determinations. As with any variation in technique, adaptations to established methods are required. This paper conveys the findings of a technical evaluation of the Polaroid film method for collecting data and presents several techniques and applications that have been developed in using a Polaroid cassette as both a Laue and a pinhole diffraction camera. Pattern interpretation is not changed and therefore is not discussed.

## GENERAL DISCUSSION

The Laue method of crystal orientation is well documented.[5] Specific requirements of this method are (1) a single crystal specimen (which could be a particular grain within a polycrystalline material), (2) a heterogeneous source of x rays, (3) a known specimen-to-film distance, and (4) fiducial marks that locate the center of the film and can be related to specimen orientation. The orientation accuracy is generally $\pm 1°$.

Laue spot diffraction from any set of planes (hkl) is accomplished by selecting a wavelength from the continuous spectrum which satisfies the Bragg law. As the integrated intensity of the continuous spectrum is approximately proportional to tube current and to the square of tube voltage, rapid exposure can be obtained by operation at maximum power ratings for the x-ray tube employed. In many cases, however, such conditions adversely affect contrast and resolution, thus reducing pattern quality.

Contrast (or spot-to-background density ratio) is always high for transmission geometries. In back reflection, however, specimen fluorescence, Compton modified scattering and temperature diffuse scattering (which all contribute to background noise) are maximized at $2\theta = 180°$, and there is consequently less contrast. In cases where considerable specimen fluorescence occurs, it is generally necessary to compromise contrast and exposure time. Resolution depends on many factors, including collimator aperture sizes and separation, specimen-to-film distance, film grain size, and whether single or double emulsion film and/or an intensifying screen are used. The appropriate experimental arrangement, therefore, is one which fulfills the specific analysis requirements.

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[2] H. K. Herglotz, Rev. Sci. Instr. 34, 708 (1963).
[3] A. E. Spakowski, Rev. Sci. Instr. 34, 930 (1963).
[4] P. H. Schmidt and E. G. Spencer, Rev. Sci. Instr. 35, 957 (1964).
[5] E. A. Wood, *Crystal Orientation Manual* (Columbia University Press, New York, 1963).